

THE IONIZATION OF GASES

BY ATOM BOMBARDMENT

Thesis by

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Abstract

A series of experiments on the ionization of noble gases by noble gas atoms is described. The results obtained are the following:

1. Preliminary results by Beeck showing little velocity dependence for ionization of argon by argon atoms between 50 and 120 volts, and showing an ionization of neon by argon atoms in this energy range are found to be due to secondary effects, principally due to argon radiation.
2. Positive evidence for the ionization of one noble gas by atoms of another is given for argon atoms bombarding neon, krypton, and xenon.
3. The inset potentials for ionization of the above combinations are measured, and are believed to be comparable in accuracy to those measured by other methods, since the measured value for argon in argon (48 volts) agrees with that of the best previous work. The values found are: argon in neon, approximately 130 volts; argon in krypton, 50 volts; argon in xenon, 55 volts.

Introduction

The ionization of gases by neutral atoms has been a subject of study only for the past six years. At a meeting of the A.A.A.S. at the University of California at Los Angeles in December of 1930, Dr. F. Zwicky¹ pointed out that on purely classical considerations, one would expect that the ionizing power of a particle increases with decreasing charge if mass and kinetic energy are held the same. Consequently if we were to allow A, K^+ , and Ca^{++} of the same kinetic energy to impinge on argon atoms, the ionizing power must decrease as the net charge increases. Since a method of obtaining an intense beam of Ca^{++} has not yet been discovered, the only way to check this theory was to make a comparison between the efficiencies of A and K^+ .

Dr. Otto Beeck² and others had already made a rather careful set of investigations as to the ionizing power, and the energy required to cause ionization to set in--the "inset potential"--for most of the possible combinations of alkali ions in the noble gases. Consequently it required only a quantitative study of the ionization of one of the noble gases by its own atoms to give a preliminary check on the theory. Argon had proved to be the easiest of the commoner noble gases to handle for positive ion work, so it was chosen for the preliminary work on atom bombardment.

The possibility of the production of a beam of accelerated atoms had been suggested by work by Kallman and Rosen^{3,4}. They discovered that the probability of charge transfer is very great for gas ions moving in a gas of their own kind, and that the neutralization of the ions

takes place without altering the translatory energy of the moving ion. Consequently it is possible to obtain homogeneous beams of atoms at energies considerably above those available by gas-kinetic methods. A sketch of the method used by Beeck^{4a} is given in Figure 1. Argon ions were formed by electron impact in chamber I. These ions were accelerated between I and II to give the desired energy. In chamber II, in which a pressure of argon was maintained, a portion of the ion beam would be neutralised. A retarding field between II and III kept positive ions from reaching chamber III, and the condenser between these chambers was supposed to sweep out secondary electrons that might have been in the beam at that point. The ionization was measured by collecting the electrons formed onto the center plate of a guard ring condenser, which was connected to the string of a string electrometer. Beeck reported exceptionally intense ionization in the case of both argon and neon when bombarded by argon atoms. In the case of argon, he observed approximately equal numbers of "positives" and "negatives", with negatives predominating slightly; but in the case of neon he observed very few positives. He also found that the efficiency of the effect changed very little between 50 and 120 volts. It is significant to note here that these effects can be explained on the basis of secondary emission, the experimental evidence for which will be described on page 25.

Encouraged by his apparent success, Beeck^{5,6} proceeded to construct a more elaborate, and more carefully designed, apparatus, which would allow the intensity of the atom beam to be measured, so that efficiencies could be computed. A schematic diagram of this apparatus is shown in Figure 2.

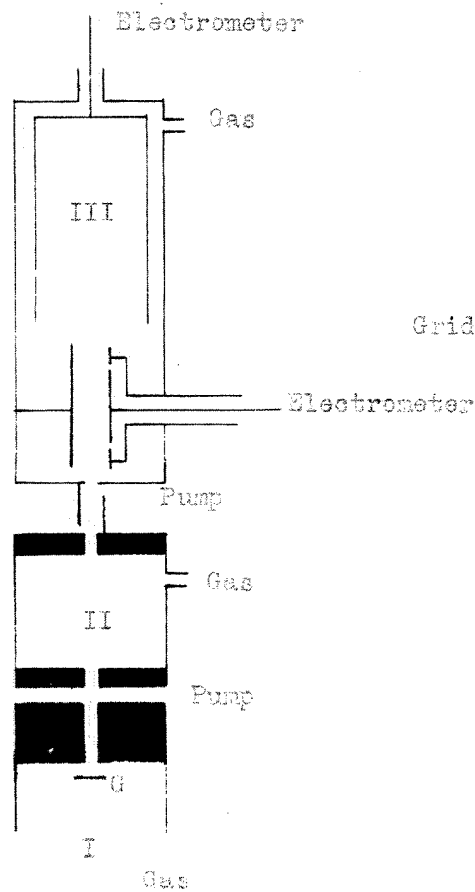


Figure 1. Beeck's Preliminary Apparatus

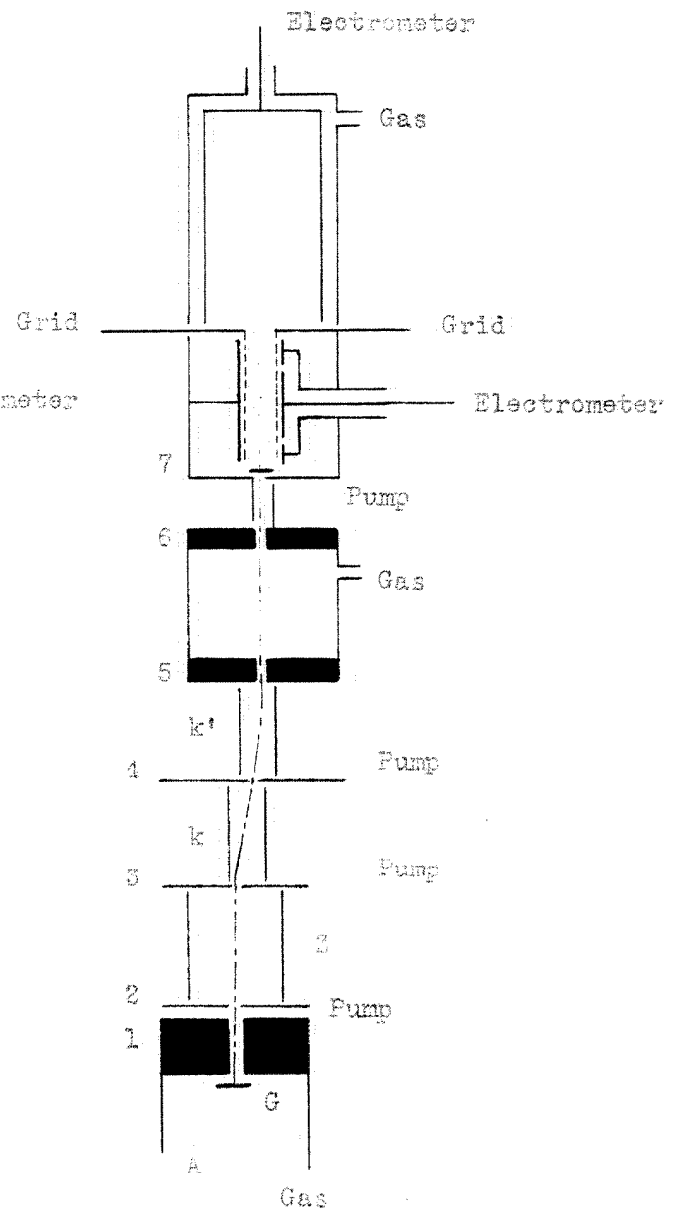


Figure 2. Beeck-Wayland Apparatus

Argon ions were formed in chamber A by electron impact. These diffused, along with the gas, through the slit 1. The ions were then picked up by the cylinder Z, which was made positive to 1 and 2, and the resulting beam homogenized by the condensers k and k'. The condensers not only gave a beam of uniform velocity, but, by deflecting the beam out of direct line of the slit 1, radiation from the discharge chamber was prevented from reaching the chamber in which measurements took place. As will be more clearly demonstrated later, the effect of the hot filament itself is small, but the effect of the radiation formed in the discharge around the hot filament can be very large.

The thermocouple was calibrated by means of an ion beam, which could be measured both thermally and electrically. Equal accommodation coefficients for neutrals and positives were then assumed in order to get a value for the intensity of the neutral beam. The principle limitation of the thermocouple was one of sensitivity--it was impossible to get sufficient intensity of atom beam to give a readable deflection from the thermocouple for energies less than 350 volts.

The ionization of the gas was measured by measuring the electrons liberated in the space between the plates of a parallel plate condenser. The actual collecting electrode, which was connected to the string of a Wulff electrometer, was protected by a guard ring to insure a uniform collecting field. The condenser plates were protected by fine grids about a millimeter from the surface, which could be made negative with respect to the condenser plates in order to hold back secondary electrons formed by impact of scattered atoms on the plates. By measuring

the intensity of the atom beam, the intensity of the ionization in the gas, and the pressure, and knowing the length of path over which the atoms had acted in ionizing the gas, it was possible to obtain the ionization efficiency of the atom beam. This was computed as the number of ionizing collisions per centimeter path per atom at a pressure of 1 mm. of mercury at 0° C. Three points were obtained: at 350 volts, 500 volts, and 650 volts. The data are plotted, in comparison with those of Rostagni, in Figure 2.

The accuracy of the measurement of the intensity of the atom beam is certainly not very great for the 350 volt point, but for the upper points the intensity of the beam could be obtained with considerable certainty. However, in light of later work it seems that we were too optimistic in assuming that we could take the entire electron current collected by the condenser as a measure of the ionization in the chamber. The grids, after all, have a finite area, and can give an appreciable secondary emission. Furthermore, although the transverse field between slits 6 and 7 would turn any secondary electrons already in the beam to the left-hand condenser plate, yet secondaries formed at slit 7 would reach the final chamber; and what is worse, the ion beam, which is deflected toward the right-hand plate, would strike that plate, knocking off electrons which would be picked up by the field and carried into the final chamber. Consequently the probability is that these results are somewhat too high.

At about the same time as this work was being done, Brasefield⁷, apparently started off by Beeck's early work, attempted to measure the efficiency of ionization of neutral atoms in various gases. His first

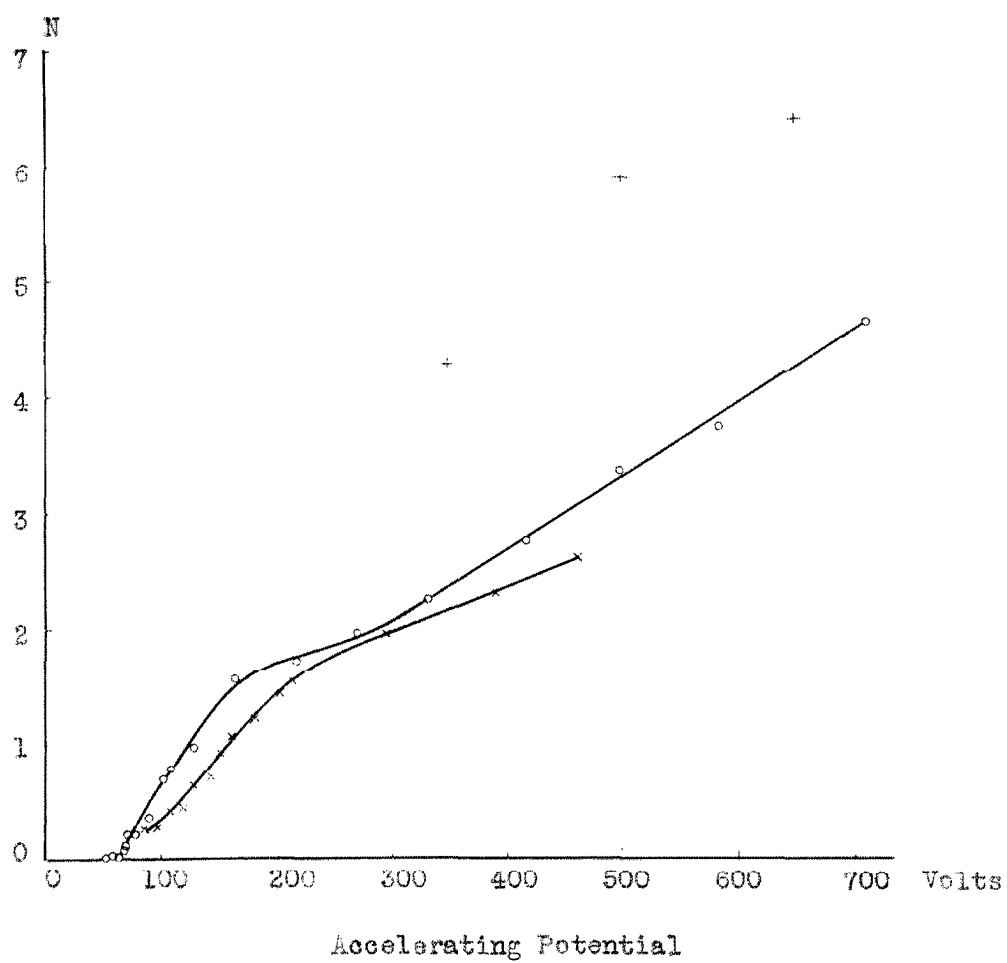


Figure 3. Comparison of Rostagni's curves with points obtained by Beeck and Wayland
 Legend: x Rostagni, Helium in Helium
 o Rostagni, Argon in Argon
 † Beeck-Wayland, Argon in Argon

work was done in a tube which was sealed off from the pumping system, so that it was useful only for measuring the effect of atoms in their own gas. The initial ion beam was accelerated by the same field which maintained the hot-cathode discharge, hence the homogeneity was bad on this account. Furthermore, no precautions were taken to eliminate, or correct for, the effects due to radiation coming from the discharge directly into his measuring chamber. Also his measuring instruments--galvanometers--did not have sufficient sensitivity to work at pressures low enough for the results to have quantitative significance. Quantitative work requires low pressures in order that each atom partake in no more than one collision, on the average.

In a second paper⁸, Brasefield repudiated most of the results of the earlier paper. In this case he worked with an apparatus similar in design to Beeck's preliminary apparatus, but again accelerated his ion beam directly in the discharge, and again used galvanometers. The intensity of the neutral beam was calculated from a knowledge of the neutralisation cross section, and from the intensity of the ion beam coming from the discharge tube. Again his results have no quantitative significance, but this time he worked over the various combinations possible with argon, neon and helium, and found no measurable effect except in the case of the gas in its own kind. This tended to throw some doubt on Beeck's results with argon in neon, though Brasefield's sensitivity was far less than that of Beeck.

In his third paper⁹, Brasefield attempted to evaluate some of the secondary effects. He was this time working with a vacuum tube electro-

meter, which gave him ample sensitivity to do some precise work. Again, however, he failed to take account of the radiation formed in his discharge tube. He allowed a mixed beam of ions, atoms, and photons, to strike a plate protected by a grid. By varying the potential on the grid, he could measure the emission of secondary electrons from the surface of the electrode. Then he put on a transverse electrostatic field, which swept the ions out of the beam. The secondary emission which was left was attributed to the neutrals, and hence they were assigned an efficiency very much greater than that possessed by positives for this process. Actually, as shown by Rostagni,^{10,12} the coefficients of emission for argon atoms and for argon ions are approximately the same, and the experimental work described on page 23 leaves little doubt that the principle effect measured by Brasefield was due to argon radiation formed in the discharge. Radiation from the hot filament could not possibly account for such a difference.

In a series of five papers^{10,11,12,13,14}, Rostagni describes a very elaborate series of experiments on the processes connected with any study of neutral rays. He made careful measurements of the efficiencies of ions and of their corresponding neutrals for emission of secondary electrons from metal surfaces for argon, neon, and helium, finding that above 100 volts the values for ions differed but little from those for atoms. Below 100 volts the coefficient of emission for atoms drops off, becoming unmeasurable below 20 volts; while for ions it remains of the same order of magnitude down to the lowest point he measured--25 volts in argon, 11 volts in neon, and 6 volts in helium. He found that the neutralisation of ions by striking a metal surface at grazing incidence was at

best a very inefficient process. He made careful measurements of the neutralisation cross sections for argon, neon, and helium positives in their own gases, finding that the neutralisation cross section increases to 20 volts, remains constant between 20 and 30, and then decreases gradually on out to 900 volts. With the assistance of all of these measurements of accompanying phenomena, he made a careful study of the efficiency of ionization for helium atoms in helium, and for argon atoms in argon. His apparatus was similar to that described above, except that he deflected his beam through 30° before neutralisation. This gave him an homogeneous beam, and at the same time kept radiation from reaching the measuring part of the apparatus. He determined the intensity of his atom beam by measuring the number of secondary electrons knocked out of a metal surface by it. He also checked his intensity by absorption measurements on the initial positive beam. He was able to take the secondary emission into account better than was done by Beeck and the author, so that his results are probably quantitatively better than ours. Also, he was able to push down to much lower velocities than we were able to do with our thermocouple. The shape of his curves--which are shown in Figure ³ along with our data--seems a bit strange in light of the curves obtained for positive ions in argon; but further experimental work, preferably using another method of measuring the intensity of the atom beam, if such can be devised, will be necessary to check this point.

In the mean time the author was working on an apparatus which would permit a measurement of the inset potential for ionization of the noble gases

by bombardment by their own atoms and of other noble gas atoms. The experiment was undertaken with the hope of eventually refining the apparatus to the point that it would be possible to measure any difference that might occur in the inset potentials for the combinations $\text{Ne}^{20}\text{-Ne}^{20}$ and $\text{Ne}^{20}\text{-Ne}^{22}$. Since, on the classical picture¹, if one requires the conservation of momentum to be satisfied by the two massive particles before and after collision (neglecting the momentum of the electron liberated), there will be enough energy available to produce ionization only when the energy of the bombarding particle is considerably greater than the electron ionization potential. The maximum energy available--for direct impact--is equal to $m_1 E / (m_1 + m_2)$ where m_1 is the mass of the atom struck, m_2 the mass of the bombarding atom, and E the incident energy of the bombarding atom. In the case for which $m_1 = m_2$, we see that the maximum available energy on this theory is 0.5 the initial energy. In the case of neon²⁰ in neon²² the available energy would be only 0.476 of the initial energy. Hence the inset should be, on this theory, about five percent higher in the second case. Such an experiment would give us the possibility of checking just how good this simple picture of the process is.

In order to insure an homogeneous beam, and to remove radiation effects from the measuring chamber, an electrostatic deflection method was tried. The ion beam was deflected through $127^\circ 17'$ by means of a cylindrical condenser. This gives not only an energy separation, but also focusses ions of given energy but slightly divergent direction¹⁵. The ion beam was then neutralised, the ions removed by a retarding field, and the resulting homogeneous atom beam used for the experiments. The ionization of the gas was to be detected by a balanced space charge detector, as

devised by Lawrence and Edlefsen¹⁶, and revised by Varney¹⁷ for similar measurements on the inset potentials for positive alkali ions in the noble gases. The principle of the device is the following: a straight tungsten filament is run down the axis of a metal cylinder. The tungsten filament is heated electrically, and a small potential difference is applied between the filament and the cylinder. If the electron current is space-charge limited, the introduction of another electron by ionization of any gas in the cylinder will not affect the current; but if a positive ion appears in the cylinder, it will be attracted toward the center. Since, in general, it will have some component of velocity perpendicular to the plane defined by its position and the filament, the atom will, in general, spiral around the filament many times before it finally reaches it. During this comparatively long time, it will considerably influence the space charge--sufficient, in fact, to neutralise, on the average, the effect of 30,000 to 40,000 electrons. This will change the current flowing between the filament and the cylinder quite considerably. If two of these chambers are balanced in the arms of a bridge, so that fluctuations of heating current and accelerating potential are pretty well balanced out, and the ionization which is to be measured allowed to occur in only one of the chambers, a very sensitive device is at hand for the detection of the positive ions formed in the process. This has many advantages over the use of electrometers or sensitive galvanometers, since it is relatively insensitive to secondary electrons. Varney had measured the inset potentials for the alkali ions in the noble gases with such a device with considerable success. However, the author found that his own apparatus--which was enclosed in a metal chamber, and consequently could not be baked out--was so unsteady that with

the intensities available for the neutral beam, it was impossible to get quantitative results. The most intense beam of neon ions to reach the upper chamber was only of the order of 10^{-11} amperes, and the neutral beam was even more feeble. Even sending the beam directly into the space-charge measuring device, eliminating the velocity filter altogether, did not give usable results with neon in neon--the only case tried with this apparatus.

In the mean time Varney^{17a} had applied his space charge apparatus to the measurement of the inset potentials of neutral atoms in their own gas. He used an apparatus (Figure ⁴§) of much simpler design than that employed by the author, having only two narrow slits to cut down the intensity of the beam, and having the apparatus sealed in glass so that it was possible to bake out the space-charge cylinders, which very materially cut down the unsteadiness of the apparatus. His values are shown in table 1, page 42.

Next he set out to construct an apparatus which would allow him to use differential pumping, so that he could try using different gases for projectiles than used for targets. His apparatus was very similar in construction to Beeck's original apparatus, except he used the space-charge detector instead of the collecting condenser. In this case photo electrons would cause no serious consequences, since the detector was sensitive particularly to positives. However, the extra slits so decreased his intensity, that he was barely able to get a measurable effect for argon in argon--the easiest case--and could get no positive results whatsoever for argon in krypton, and argon in xenon. Even for

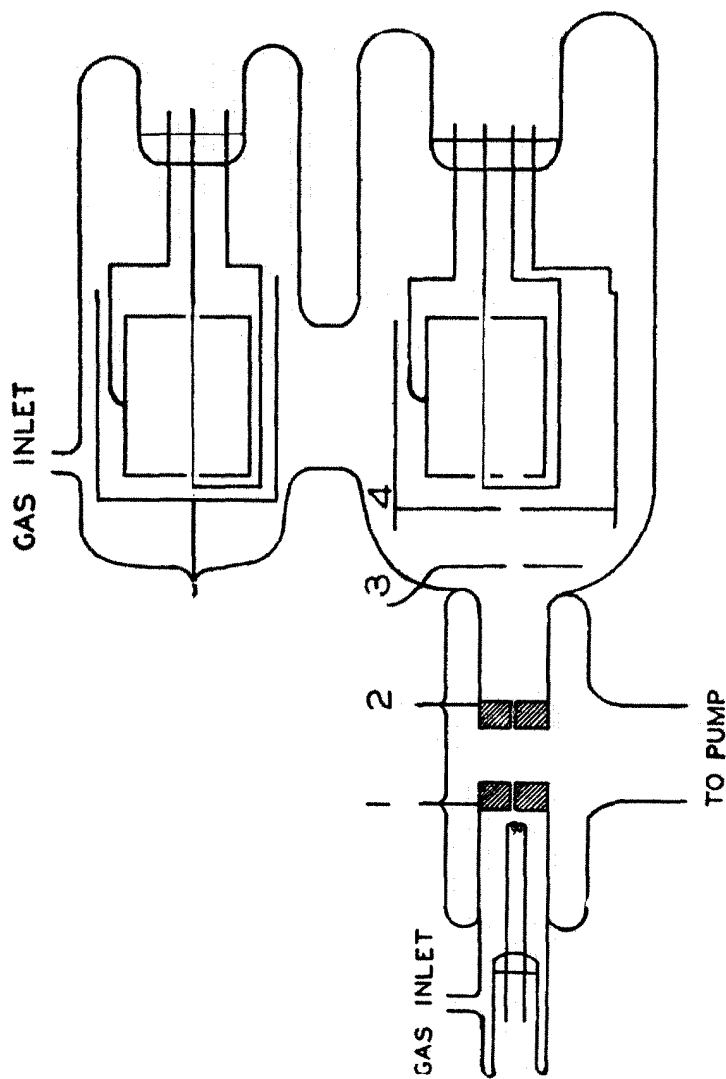


FIG. 1. Diagram of tube.

Varney's Apparatus

the argon-argon case, he could come no closer than within ten percent of his earlier results.

Varney's difficulties in the "mixed" cases, coupled with the fact that circumstances forced him to abandon the field, made it seem feasible for the author to go ahead and attempt to measure at least some of these cases by another method, always with the hope that sufficient sensitivity could be obtained to allow a measurement in the cases of the neon isotopes. It was also important to try to clear up the discrepancies between the works of various experimenters in the field. Particularly to account for Beeck's preliminary results, showing ionization of neon by argon, and showing practically no velocity dependence of the ionization of argon by argon; and to account for the discrepancy between Brasefield's and Rostagni's values for the coefficient for emission of electrons from metal surfaces by atom bombardment.

Description of Apparatus

1. Production of the ion beam

The ion beam was produced by electron bombardment in chamber I. (Figure ⁵4)

The electrons were emitted by a heated tungsten filament and accelerated toward slit 1 by a field applied between the filament and a grid G. The filaments used throughout all of this work consisted of spirals wound from 8 mil pure tungsten wire on a 20 mil piano wire mandrel. 5 mil piano wire was wound simultaneously with the tungsten, but held from the opposite side in order to keep the mandrel, which was turned from both ends in a small winding machine, straight. A spiral about three inches long would be wound, and then mandrel, spacer, and tungsten spiral were all placed in concentrated hydrochloric acid. This dissolved out the iron mandrel and spacer, leaving a very uniform tungsten spiral. This was cut in 1 cm. lengths to be used for filaments. These were mounted with nickel leads made from 30 mil nickel wire, the end of which was filed down in a lathe so that it would fit tightly into the end of the spiral. The spiral was spot-welded to the nickel lead. This filament assembly was then welded across heavy nickel leads, attached to tungsten-pyrex seals through a ground joint which permitted the filament assembly to be removed for replacing the filaments. The grid was made by winding 5 mil tungsten wire on an 8-32 machine screw, cutting the resulting spiral to the right length, and welding it along one side to a 20 mil nickel wire. This, in turn, was welded to a support sealed through the ground joint. This allowed the possibility of applying a different potential to the grid than was applied between the filament and slit 1, although this degree of freedom eventually was found unnecessary, as maximum emission was obtained when the entire

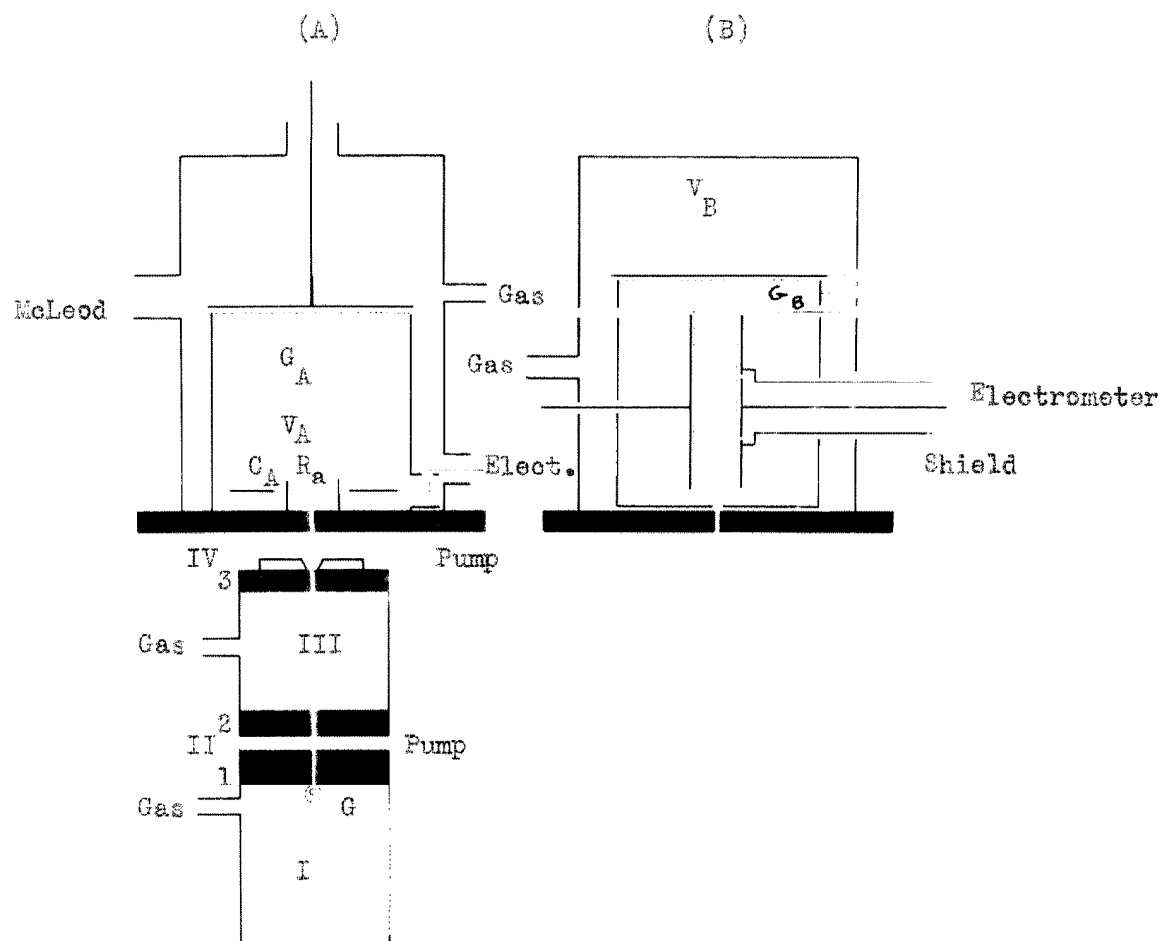


Figure 5. Apparatus used in this work

electron accelerating potential was applied between the filament and grid. The grid and filament were not mounted concentrically, the filament being mounted closer to the side of the grid which was just in front of slit 1.

Slits 1, 2, 3, and 4 were all 0.5 mm. wide and 5 mm. long. Slit 1 was 8 mm. deep, while slits 2, 3, and 4 were 5 mm. deep. A high speed mercury diffusion pump was connected to chamber II, so that there would be little neutralisation of the ions emerging from 1 in their passage between 1 and 2. This precaution was considered to be necessary, since no other device was used to homogenise the beam. Certainly the ions emerging from slit 1 would have only at most kinetic velocities, when no potential was applied between 1 and 2, since any fields applied in chamber I would have pulled them towards the filament.

2. Production of the Atom Beam

The ions emerging from I were accelerated by an electric field between 1 and 2. This accelerated beam of ions then entered the "neutralisation chamber" III. In this chamber a gas pressure could be maintained, allowing a part of the ion beam to be neutralised by the process mentioned in the introduction. From slit 3 there emerged a mixed beam of positives and neutrals, from which the positives could be removed by applying a retarding potential to slit 4. Slit 4 was either connected directly to 1, so that the retarding field was equal to the accelerating field, or an additional 22 1/2 volt battery could be connected in to make absolutely sure that no positives would reach the final chamber. Although such a field removes the positives effectively enough, it acts as an acceler-

ating field for any electrons that might be in the beam due to secondary emission at various metal surfaces struck along the way. Consequently, a small permanent magnet was placed just above slit 3, in order to deflect any electrons out of the beam.

3. Measurement of ionization

Two different measuring chambers were used. The first (V_A) turned out to be useful only in determining the origin of the most important secondary effects. This arrangement was a modification of that used by Sutton, Mouzon, and Beeck² in their work on the ionization of gases by positive ion bombardment. A collecting ring C_A , protected by a guard ring R_A from impacts by the direct beam, was made positive with respect to the grid G_A . The field between the collecting ring and the grid brought any electrons formed in the space between to the collector, but it also collected secondary electrons emitted from the grid and from the walls of the inner chamber, which was at the same potential as the grid. Electrical connection to the collecting plate was made through brass rod, insulated from the metal chamber by amber bushings, and made vacuum tight with ceresin wax on the outside.

The final measurements were taken with collecting head V_B . In this apparatus a parallel plate collecting arrangement was used. In this case the shape of the field in the neighborhood of the actual collecting plate was much more uniform than with the preceding apparatus, and in addition all metal surfaces were protected by grids. The grids protecting the condenser plates were made of 2 mil nickel wire soldered to narrow brass frames. The wires were mounted 1 mm. apart, and the front of the grid was 1 mm. from the plate it covered. The grids were supported by pins extending from the

four corners and passing through small insulating bushings of Bakelite BT 61, which protruded 0.5 mm. beyond the front surface of the condenser plates. This 0.5 mm. plus the thickness of the brass frame, which was the same, gave the spacing of the grid from the condenser plate.

The actual collecting plate P_E was supported by a brass rod, which also formed the electrical connection to the outside, passing through a single amber bushing. This connection was made vacuum-tight with ceresin wax on the outside of the apparatus. The brass rod supporting the central part of the condenser arrangement was surrounded by a brass tube which formed the support for the guard ring, and also formed an electrostatic shield for the electrometer lead. This tube was led out of the vacuum chamber through a redmanol bushing, and was electrically connected to the case of the electrometer through the continuation of the electrostatic shield.

The condenser arrangement was supported in a chamber inside of the vacuum chamber, which could be maintained at a different potential from the main chamber to keep secondary electrons formed on the walls of the outer chamber from reaching the collecting plates. A slit was placed in the bottom of this inner chamber. When the inner chamber was made negative with respect to the outer, electrons knocked off of slit 4 by the atom beam could be prevented from reaching the measuring chamber. The inner chamber was supported on a ring of Bakelite BT 61, so that slits 4 and 5 were only 1 mm. apart. The electrical connections to the grids, condenser plates, and inner chamber were taken out through redmanol bushings.

Above the grid G_B two different arrangements were used: the first consisted

of a flat plate placed 1 mm. above the grid; the second merely did away with this plate, allowing the beam to strike the top of the vacuum chamber. No significant difference could be obtained by changing from one to the other, so most measurements were made with the plate removed.

All surfaces except the grids and their frames were blackened with benzol soot. The grids and grid frames were blackened with Aquadag--colloidal graphite in aqueous solution. This was to cut down secondary emission by atom bombardment, and to cut down reflection of radiation from the various metal parts. The grids could not be sooted, as the heat of the flame was sufficient to warp them.

4. Electrical measurements

The electrometer used for the measurement of the electron current was a Wulff string electrometer, built for Beeck's work with positives in the noble gases. This had a maximum sensitivity of 1000 scale divisions per volt, though was usually used at about half this sensitivity. The capacity of the electrometer circuit was calculated as approximately 50 centimeters. At full sensitivity, allowing 40 seconds for a traverse of 2 scale divisions, a current of 3×10^{-15} amperes could be measured. Actually, readings were kept below 20 seconds whenever possible, and a larger traverse--at least 5 scale divisions--was usually used. The accelerating potential was measured by a Siemens-Halske volt meter, 300 ohms per volt. This meter was calibrated against a Weston standard cell by means of a Brooks deflection potentiometer with volt box made by Leeds and Northrup. The standard cell was calibrated by Dr. Dunnington against cells whose values were known. The meter was found to read 0.3 volt high in the region between 40 and 60

volts. The insets for argon in argon, krypton, and xenon were found to be in this region. This correction was negligible for the overall accuracy of the measurements. In the region between 100 and 150 volts both scales used were found to be between 0.8 and 1.1 volts high. This also is negligible for the accuracy of the measurements made in this region. The electron acceleration and ion acceleration potentials were supplied by a bank of Exide 6 CTM storage batteries. These batteries have a low capacity, about 15 ampere hours, but proved to be very steady for the small drains used in this work. Other potentials were furnished by radio "B" batteries.

5. Method of handling gases

The gases were admitted to the various chambers through Smythe leaks¹⁸ from 300 cc. gasometer bulbs for argon and neon. The gasometer bulbs were filled from liter flasks of the gas obtained from the Air Reduction Sales Company. The gases were bought in pure form, and no further attempt to purify them was made. However, between the leaks and the apparatus, a liquid air trap was placed, in order to remove grease vapors and mercury vapor from the gases. The krypton and xenon were left over from bulbs brought by Beeck from Germany--a gift to him from the Linde works. Since the gases had been sealed in by stopcocks for several years, their purity was tested spectroscopically before they were used. The only contamination found was mercury vapor, which the trapping system did not completely remove from the discharge tube. These gases were passed through a CO₂ trap before being used, as liquid air would condense them out.

The pressures used in chambers I and III were measured by vibration gauges. Such a gauge consists of a flat quartz fiber about 0.2 mm. by 1 mm. and a-

bout 6 cm. long. One end has a slight globule melted on, the size depending on the pressure range in which the gauge is to operate. The other end is fastened to a projection from the wall of a tube, so that the fiber hangs vertically downward. A magnetically operated hammer allows the fiber to be set into vibration, and the time for the vibration to damp down to half amplitude is measured. These gauges can be calibrated against a McLeod to give very reproducible results in a 50 fold pressure range. In this apparatus the readings of the pressure gauges which corresponded to maximum emission were obtained empirically, and used in adjusting the leaks, without any attempt at obtaining the absolute value of the pressure. In the measuring chamber, however, the pressure was measured on a McLeod gauge calibrated by Beeck. The McLeod was connected to the chamber through a trap which was cooled by liquid air for work with argon and neon, and with a mixture of alcohol and solid CO_2 for krypton and xenon.

When the gas in the top chamber differed from that used for producing the atom beam, it was impossible to use a sufficiently high pressure in the neutralising chamber to insure the maximum efficiency of neutralisation, since the interdiffusion would contaminate the gas being bombarded. Consequently, the pressure in the neutralisation chamber was adjusted to such a value that the pressure in the upper chamber was at most 2×10^{-4} mm. of Hg, when no gas was admitted directly to that chamber. Since the lowest pressures used in the upper chamber were 2×10^{-3} mm. of Hg, the contamination was kept to 10 percent or less.

Measurements

1. Collecting head A

This arrangement proved to be useful particularly in locating the exact nature of the most troublesome of the sources of secondary electrons which tended to mask the measurement of the actual ionization in the gas. As will be noted from a look at the diagram of the apparatus, only the surface which the direct beam of atoms struck was protected by a grid. Consequently any secondary electrons knocked off the side walls by scattered atoms or by scattered radiation would be captured by the collector. The most disconcerting thing in attempting to make measurements with argon atoms in argon was the copious emission of electrons that took place for atom beams with energies even below the value found by Varney for the inset. This emission seemed to be practically independent of the presence or absence of the residual positive ions in the beam, even when the positive ion current was greater than the intensity of the neutral beam could possibly have been. Consequently it was suspected that radiation must be the principle cause. The filament did not give sufficient photoelectric effect even to be measurable on the electrometer at the sensitivities used--about 500 scale divisions per volt, which allowed currents as feeble as 1×10^{-14} to be measured. However, as soon as electrons were accelerated with sufficient energy to ionize, and incidentally, also to excite, the argon in the lower chamber, the effect began to show up. Consequently it was assumed that the principle offender was the resonance radiation of the argon. To check this hypothesis, a series of measurements was taken with various conditions in the upper chamber, and with only radiation and 60 volt positives bombarding it, since chamber III was evacuated. With no gas admitted to the upper chamber, a potential of 20 volts applied to the upper plate reduced the

current collected on the collector to 16 per cent of its original value, with the plate at the same potential as the grid. With argon at a pressure of 8×10^{-3} in the upper chamber, and the same bombarding beam, twenty volts applied to the plate reduced the current collected only to 73 per cent of its value with the plate at the same potential as the grid. With neon in the upper chamber, at a pressure of 1.5×10^{-2} mm. of Hg, a potential of 20 volts on the plate reduced the current collected to 34 per cent of the initial value. The large amount of emission apparently coming from the body of the gas in the argon-argon case could not possibly have been due to ionization of the gas, because even if the beam had been neutralised in chamber III, the ionization at 60 volts had been found to be only a small fraction of the background. And in this case the neutral component must have been very small indeed. The positives could have caused no ionization, as Wolf¹⁹ has found that ionization of argon by argon ions does not set in until the ions have about 300 volts energy. Consequently the effect must be explained by scattering of the ion beam and of the argon radiation. Since cutting off the positive beam made only a small difference in the total effect measured, the radiation is left as the principle offender. The most rational explanation is that the argon radiation is absorbed by the argon, and re-emitted in all directions, knocking secondary electrons off the side walls. Neon, on the other hand, would show no such selective preference for argon radiation, and consequently would not give nearly as strong an effect from the side walls.

The fact that the argon resonance radiation is in the extreme ultraviolet makes it exceedingly effective in knocking off photoelectrons. The copious emission of these secondaries by the radiation explains Brasefield's spurious results as to the coefficient of emission of electrons by neutral

argon atoms impinging on a metal surface, as well as accounting for the apparently copious ionization of both argon and neon found by Beeck in an apparatus completely unprotected as far as photo emission was concerned. The fact that for argon in argon he measured almost as many positives as negatives is explained by the fact that, if the collecting field is such as to remove the photoelectrons from the plate connected to the electrometer, the string will be charged up positively. Consequently the only difference between "positives" and negatives will be due to the extra secondary negatives formed elsewhere. In the case of neon he would observe few "positives", because the neon would not be very efficient in scattering the radiation to the condenser plates, even at high pressures, and the pressures he used were of the order of a few times 10^{-3} mm. Hg. However, there must have been a fair number of photoelectrons emitted from the "grid" at the top of the condenser-chamber, as it was constructed of a piece of bronze window screen made of quite heavy wire.

2. Collecting head B

a. Methodology

As was mentioned in the description of the construction of this apparatus, the major surfaces were covered by grids which could be made negative with respect to the surfaces in order to hold back electrons knocked off. Consequently the first thing to do was to determine the correct potentials to apply to insure the optimum conditions.

With all of the grids connected to the plates which they covered, the effect of the collecting potential between the condenser plates was studied. This was done in the following manner: a collecting potential of 30 volts was taken as a standard. With a beam of argon atoms bombarding the chamber,

which contained argon at a pressure of a few thousandths of a mm. of mercury, the time required to charge up the center portion of the right hand plate--which was connected to the string of the electrometer--from +0.05 volts to -0.05 volts was measured. Then the collecting potential was changed to 1 1/2 volts and the time for collecting the same charge was measured. Immediately the collecting potential was returned to 30 volts, and the rate of charging up of the electrometer measured again. If the "standard" had not changed appreciably, a new collecting potential was tried, and so on, each time coming back to the 30 volt point for a standard. In working up the data, the current collected for a 30 volt collecting potential was taken as unity, and the other values compared to it. An average of the 30 volt readings before and after each other reading was used in computing the ratios. The current collected at 30 volts tended to drop with time, due to the gradual decrease in pressure in the various chambers as the pressure in the gasometer bulbs dropped: the slits in this apparatus were, after all, pretty large, and consequently it used quite a lot of gas. "Saturation" curves for grids at 0, 6 and 12 volts negative with respect to the condenser plates are shown in Figure 6. The 6 and 12 volt curves were taken no further than 30 volts, since in general it was deemed advisable to keep the collecting potential so low that the electrons accelerated by this field would not have sufficient energy to ionize the gas. The actual collecting potential used for most of the final measurements was only 12 volts, which will be seen to give nearly 90 per cent of saturation, though in no case was any significant difference found with 30 volts collecting potential. Since the purpose of the experiment was to measure inset potentials, it was more important to keep down secondary effects to a minimum than to be sure that every liberated electron was collected. The effect of varying the grid potentials with the collecting

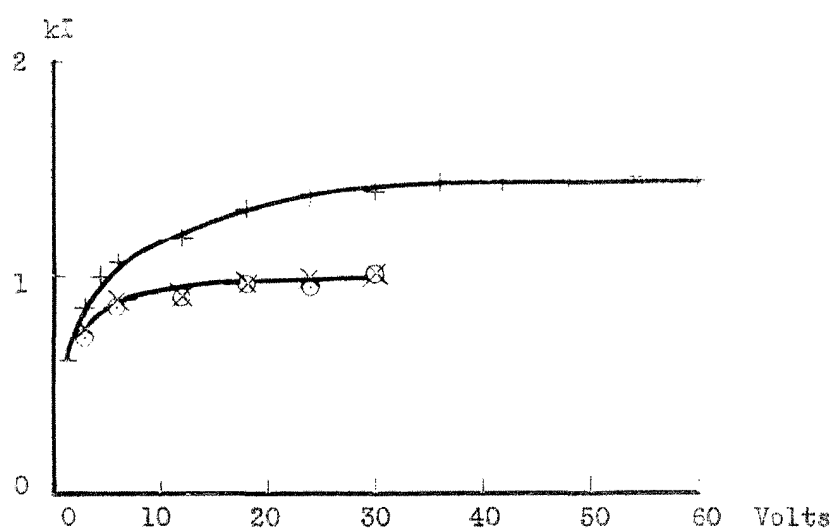


Figure 6. Collecting potential required for saturation

Legend: + grids at 0 volts
o grids at 6 volts
x grids at 12 volts

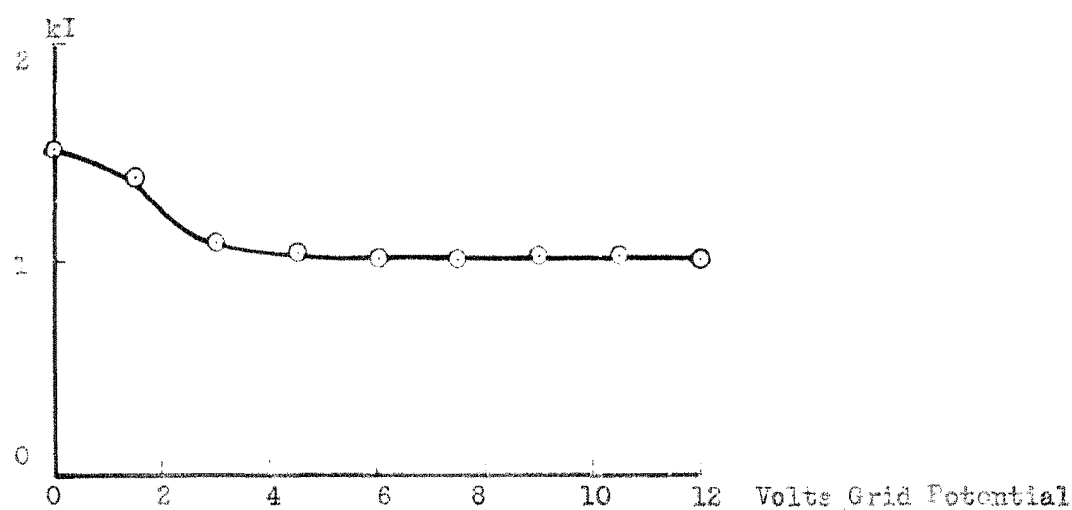


Figure 7. Effect of grids on suppressing secondaries.
Collecting potential: 30 volts

potential fixed is shown in Figure 6, hence it is clear that grid potentials between 6 and 12 volts would be sufficient to hold back the secondaries emitted from the condenser plates.

The upper plate was normally made 22 1/2 volts positive with respect to the grid, which proved sufficient to hold back the secondary electrons emitted from its surface. Here it was not considered necessary to keep the electron acceleration below the ionizing potential of the gas, since electrons formed would be pulled toward the plate, and not into the measuring space. Slit 5 was made negative with respect to slit 4, 45 volts proving sufficient to give a minimum of secondaries.

In spite of all of these precautions, secondary electrons still formed a fair percentage of the measured effects. However, with optimum conditions in the upper chamber, it was possible in several cases to get effects that seem attributable only to ionization of the gas by atom collision. Although the actual measurements were taken collecting negatives, in all cases except neon in neon a check run was made collecting positives to be sure that they were present. Since it required 60 volts collecting potential to get even close to saturation for positives, it was deemed advisable not to attempt to collect them for an actual determination of the inset potential, since the electrons, both primary and secondary, had plenty of energy to ionize the gas themselves. Any attempts to collect positives with low enough collecting potentials to assure no ionization by the electrons proved to give such small currents that no good determination of the inset could be made.

Runs were made as quickly as possible, in order to avoid changes in pressure,

and fluctuations due to warping of the filament as the supports got heated. Considerable practice was required before it was possible to get runs in which the time element was not serious. It was found to be impossible to reduce data taken at different pressures to a common basis by any method of correction that presented itself, so data were plotted run for run, and the inset potential obtained from comparing various runs. The data were taken by assuming a point, which a preliminary run showed to be below the inset as a standard. A value for this accelerating potential was taken, the accelerating potential changed, and the current to the collecting plate measured again, whereupon the accelerating potential was returned to the "standard" value. If this had not changed appreciably, the run was continued. If the current collected at the "standard" points drifted slowly downward, as would be expected from decreasing pressures, the run was used. If, however, the "standard" was erratic, or drifted very rapidly, so that interpolation was dangerous, the run was thrown out. Runs were taken with various pressures of the gas being bombarded in the final chamber, and with various conditions in the discharge tube. Values for the insets obtained for different pressures were in agreement with each other to the limits to which the curves could be read. For very low pressures the total effect of the ionization was so small that the slopes of the curves obtained were too small to allow an accurate location of the potential. Also, for low pressures, the effect of the residue of argon in the upper chamber was appreciable.

b. Argon in argon

Argon in argon was studied for two reasons: due to the comparatively high efficiency of ionization for argon neutrals in argon, it was ideal for getting used to handling the apparatus. In addition, Varney had already

measured this case by another method, and it would give a good check of the accuracy of this method. However, argon not only gave the largest ionization currents, but also the largest absolute value for the background. Although considerable care had been taken to keep down the unprotected surface to a minimum, still there was enough to furnish a considerable emission of secondaries. Although many runs were made with argon in getting used to the apparatus, very little data were taken under the conditions set up as necessary to be fulfilled for a useable run. None of the runs taken were out of line with the "useable runs", all giving an indication that ionization set in in the neighborhood of 50 volts. The two runs which fulfilled the required conditions for steadiness and speed are given in Figure 8. It will be seen that the check with Varney's value of 48 volts \pm 2 volts is excellent--so good that further work with argon in argon was deemed unnecessary. In working with argon in argon, the background was so large that the electrometer had to be used at rather low sensitivities in order that the time of traverse of 20 scale divisions could be measured with accuracy. Consequently the sensitivity for measuring ionization was considerably cut. For argon in argon, the usual charge collected was 0.1 volt, while the largest charge collected in other cases was half that, and frequently much less than half. The time required to charge up the electrometer to the required amount was always kept between 5 and 40 seconds, but in general it was adjusted to be between 15 and 20 seconds for the background, since this allowed rapid work, and at the same time considerable accuracy in handling the stop watch. The electrostatic capacity of the system was estimated to be between 30 and 50 centimeters. In general, data were plotted in percentages of the background, since that gives a good idea of the reliability, and the absolute value is of no consequence for measuring insets.

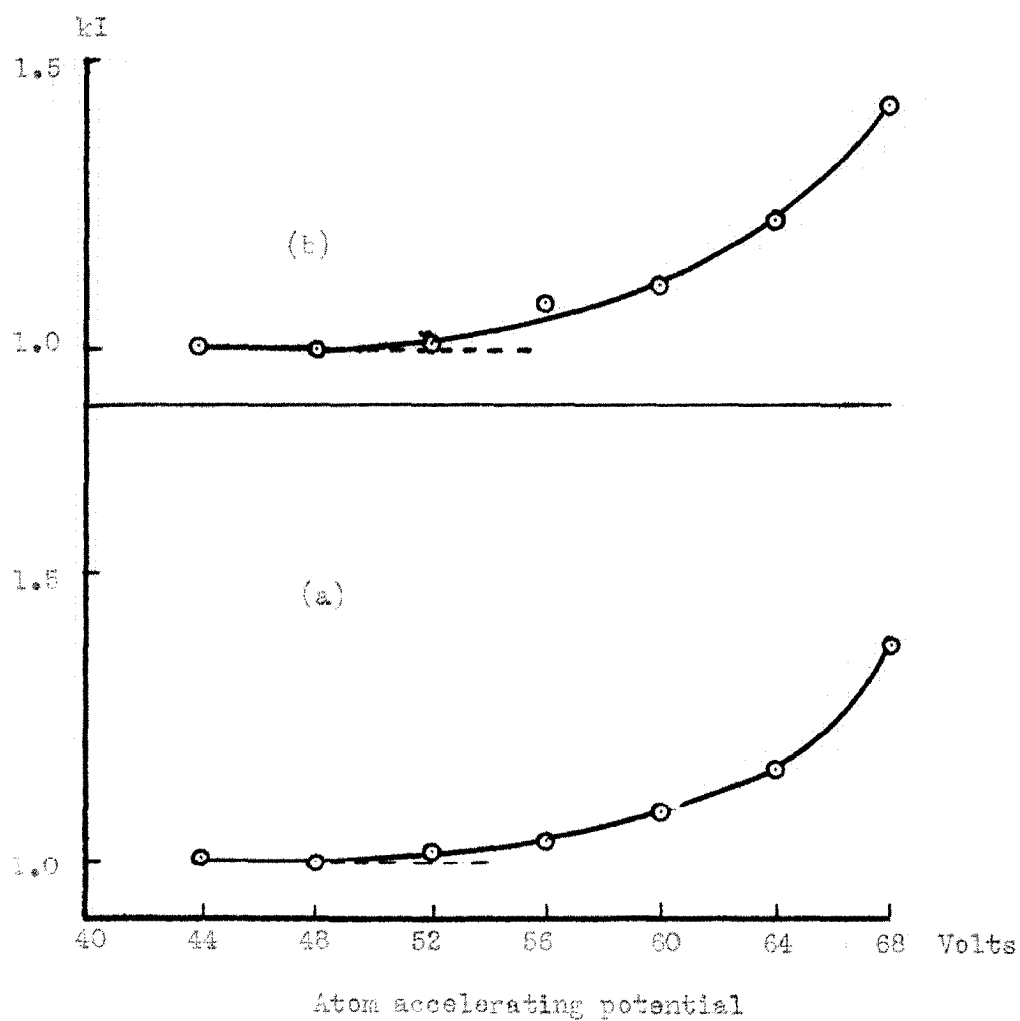


Figure 8. Ionization of Argon by Argon

c. Neon in neon

The efficiency of ionization for neon neutrals seems to be exceedingly small, and in addition it is more difficult to get an intense beam of neutrals. In the only curve obtained for this case, Figure 4, it is seen that the rise above the background is very small. The likelihood of making a measurement of the inset for neon in neon with any degree of precision with this apparatus is seen to be small. Certainly it would have been impossible to detect the small difference to be expected for the different isotopes with the small amount of neon²² available. Since Varney's result of 78 volts should be more accurately determined than anything that could be done with this apparatus, no attempt was made to do more work with neon in neon.

d. Argon in neon

The first evidence for atoms of one gas ionizing another was obtained in the case of argon atoms in neon. Curves a, b and c, Figure 10, gave promise that there was a real effect. Curve d, taken at a much higher pressure than the other curves, gives very definite evidence of ionization, and a fair indication of the inset. From these curves the inset is estimated to be in the neighborhood of 130 volts. This set of curves was taken with the collecting potential at 30 volts, grids 12 volts, and the electron acceleration in the discharge tube at 40 volts for b, c, and d, and at 50 volts for a. The pressure of neon in the upper chamber was about 4.5×10^{-5} mm. Hg for a, 9.5×10^{-5} for b, 4.5×10^{-5} for c, and 1.6×10^{-2} for d. It was found in general that the effect went up with increase in pressure, though the background also rose. However, pressures in the discharge tube and neutralisation chamber also had their effects in changing the relative intensity of radiation and atom beam. The neon data is all plotted relative

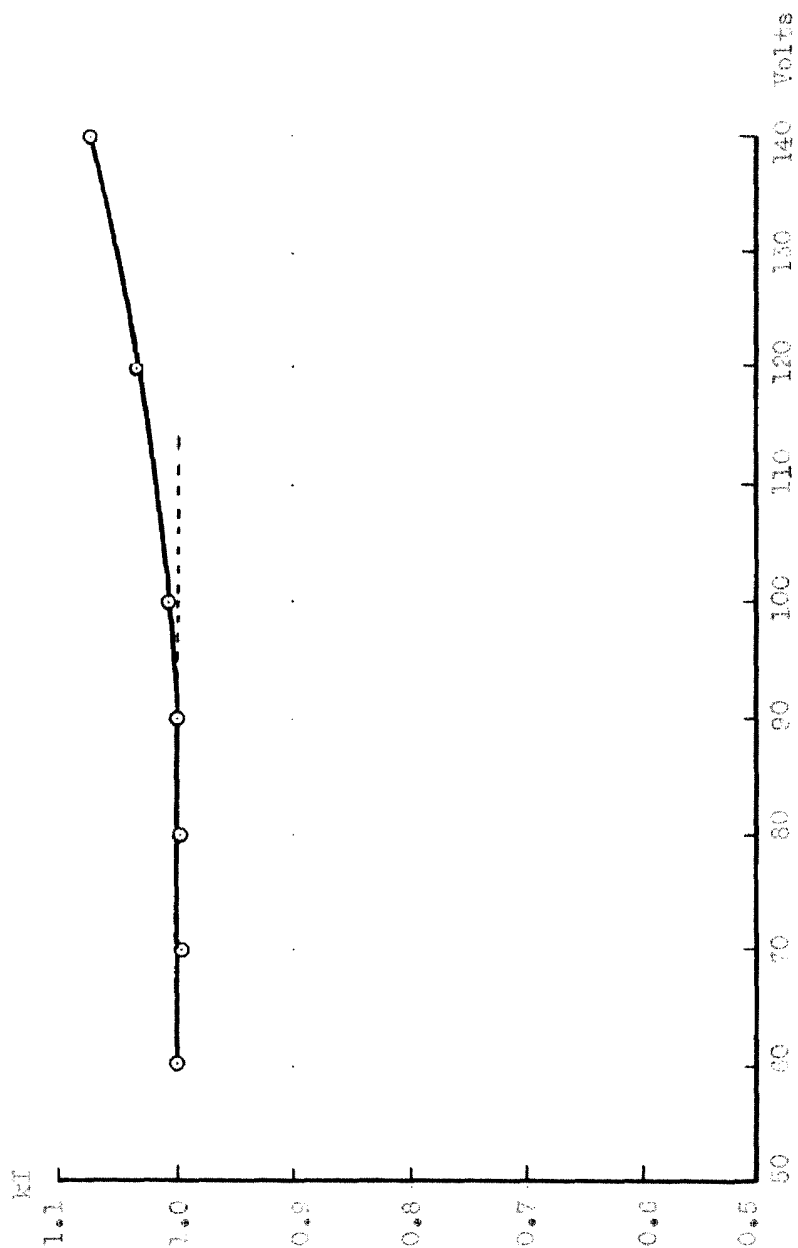


Figure 9. Ionization of Neon by Neon
Atom Acceleration Potential

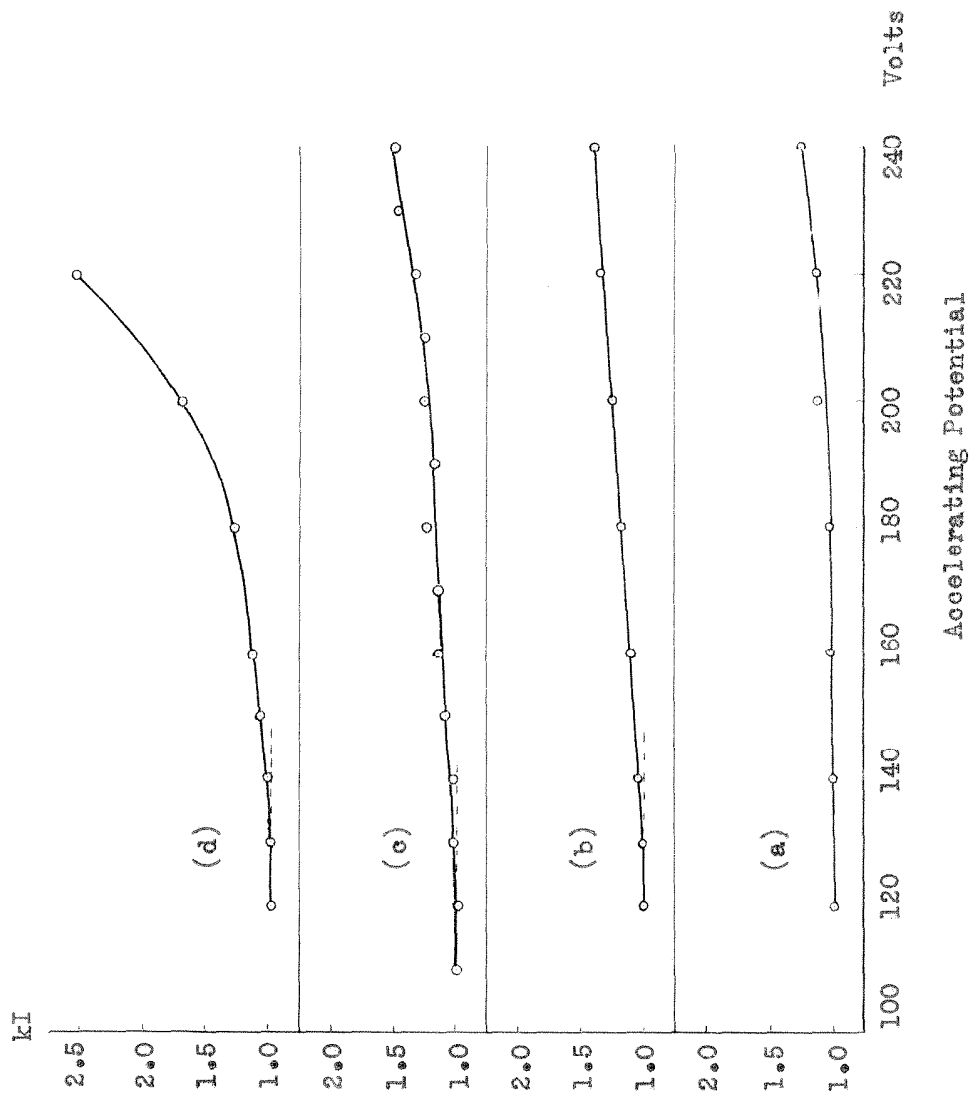


Figure 10. Ionization of Neon by Argon

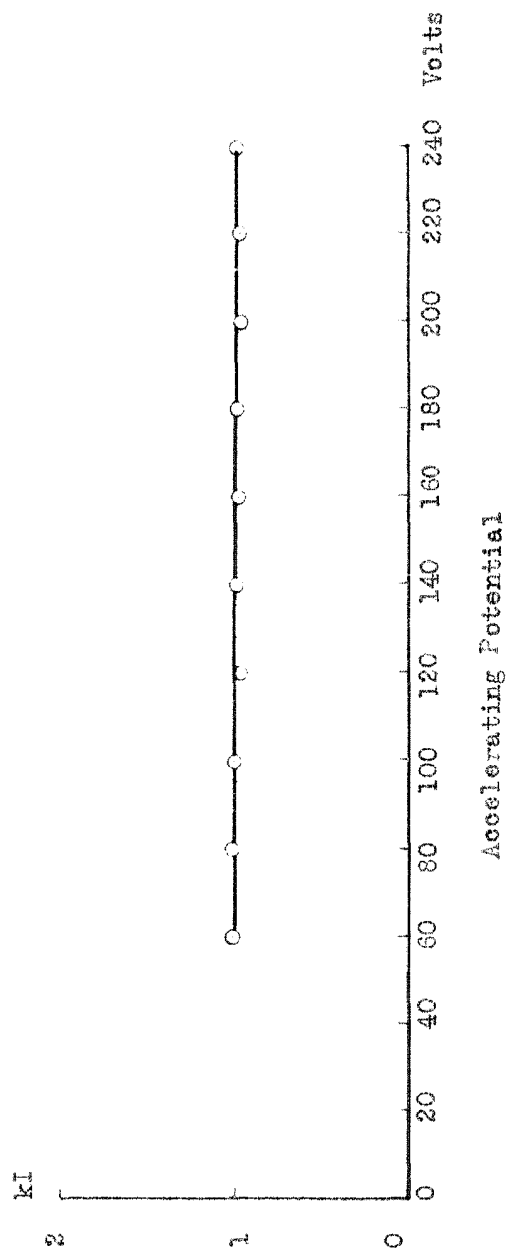


Figure 11. Background for Argon in Neon

to the background as unity. Preliminary experiments indicated that there was no evidence of ionization below 120 volts. In order to be absolutely sure that the effects measured were not due to the residual argon in the upper chamber, a run was taken over this energy range--see Figure ~~10~~¹¹--the flatness of which leads one to conclude that the effect in neon is real.

e. Argon in xenon

Due to the small amount of gas available, the number of runs which could be made for argon in xenon was distinctly limited. Figure 12 a shows the results of a preliminary run taken with 10 volt intervals. Curve 12 b was taken shortly after with 5 volt intervals in the hope of getting a better measure of the inset, but the intensity of the initial atom beam had dropped in the mean time. The xenon pressures were about 4×10^{-3} mm. Hg in case a, and 3.5×10^{-3} in case b. Trouble with the gasometer system forced the runs to be stopped, and resumed a few days later. The McLeod gauge was out of order for these runs, so the pressure was not known exactly, but probably was somewhat higher than for the earlier runs. Curve c shows the relative effects for the first of these runs. Something went wrong with the filament for the second, so a complete run could not be taken. However both gave a definite indication of a break between 55 and 60 volts, as shown so clearly in curve c. Sufficient gas was not at hand further to check these values. The collecting potential used in all of these runs was 30 volts, the grid potentials 12 volts, and the electron acceleration in the discharge, 40 volts.

f. Argon in krypton

Here again the small amount of gas available made rapid work absolutely essential. Unfortunately about half the gas had been wasted in attempting

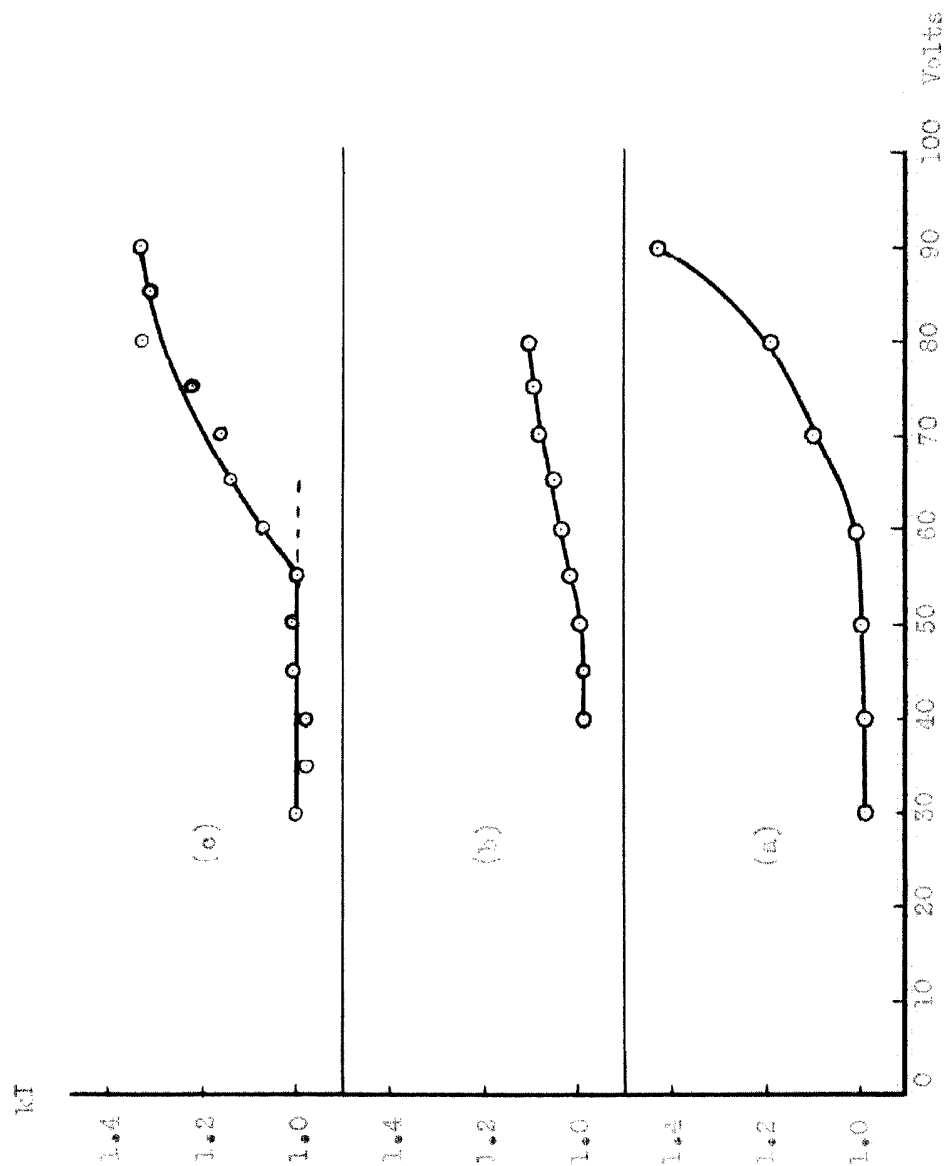


Figure 12. Ionization of Xenon by Argon

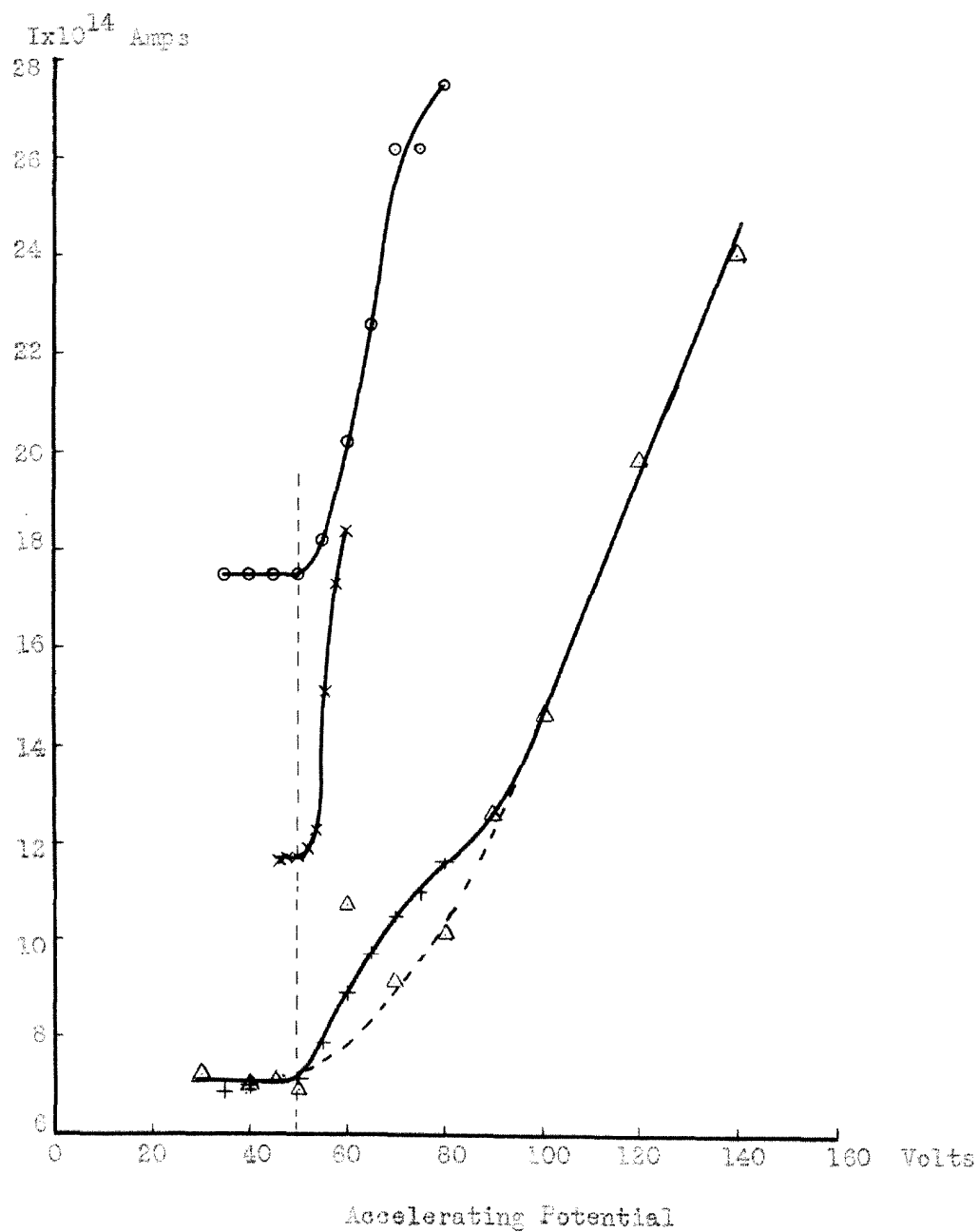


Figure 13. Ionization of Krypton by Argon

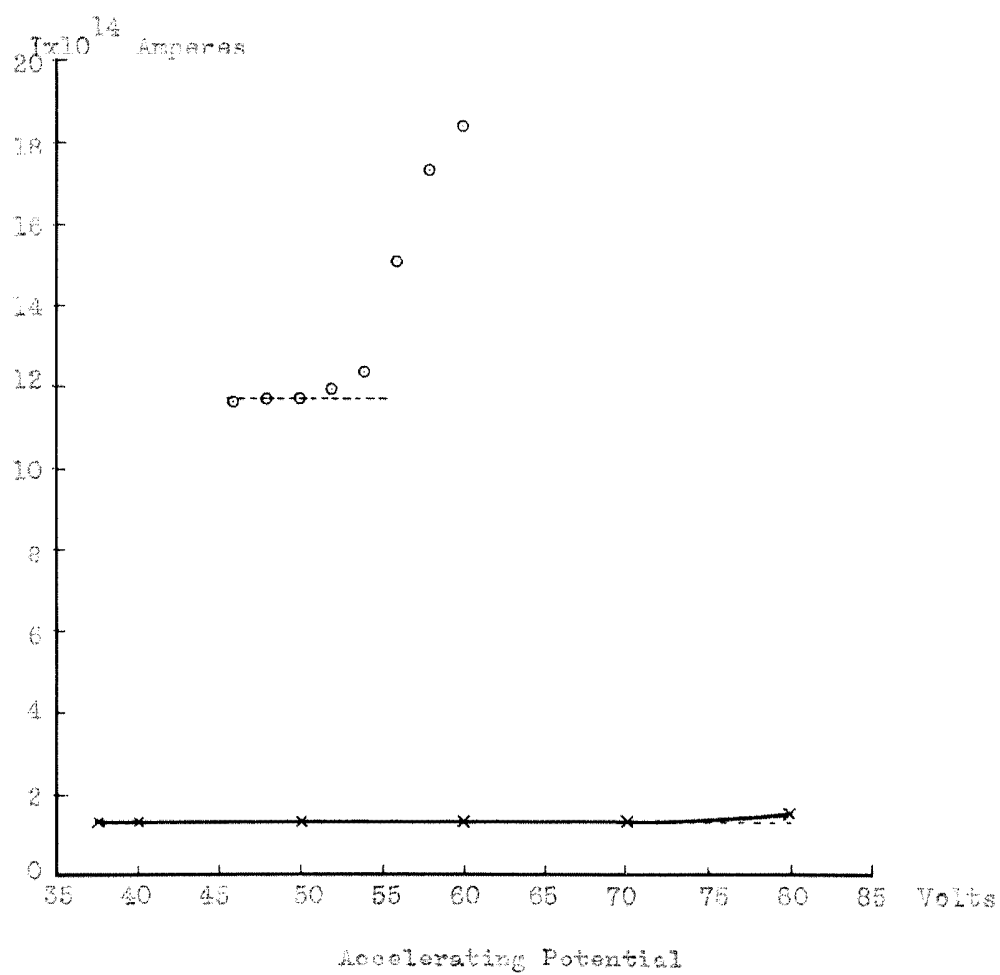


Figure 14. Background for Argon in Krypton
Legend: o Argon in Krypton
x Background

to get results with an intermittent ground in the filament circuit. However, the set of runs shown in Figure 13 more than made up for the previous difficulties. Curve a was taken hurriedly at 10 volt intervals to establish the region in which it was worth while to look for the inset. Curve b was then taken--these two curves were both taken at a rather low pressure to conserve gas, about 3.3×10^{-3} mm. of Hg--at 5 volt intervals to fix the inset more closely. Then the pressure was raised to 7×10^{-3} and curve c taken at five volt intervals. The flatness of the curve below 50 volts indicated that inset must occur at that point or above, so curve d was taken with 2 volt intervals. The pressure was beginning to fall rapidly, so no further curves were taken. All the curves are plotted on the same scale of currents collected, taking as the value for the background the average of the background readings throughout the run, and taking as the capacity of the system 5×10^{-11} farads, which was estimated to be an upper limit to the capacity. Although the major differences in background are probably due to the differences in pressure in the measuring chamber, the emission may very well have dropped some throughout the sequence of runs. Immediately after the last run, all of the gas was turned off, and the system allowed to pump down to a pressure of about 10^{-5} mm. of Hg as estimated from the McLeod, which was exceedingly insensitive in this region. Then the stopcocks leading from the leaks to chambers I and III were opened and the system allowed to come to equilibrium. The pressure in the upper chamber was found to be 2×10^{-4} mm. of Hg. The filament was then turned on and a background set of readings taken. This is plotted to scale in Figure 14 along with curve d. It is obvious from this comparison that the inset for ionization of krypton by argon in the neighborhood of 50 volts is real. Also the shape of the background curve leaves little doubt as to the reality of the effect in xenon, even though the inset is not so sharp as in krypton.

Discussion of results

The conclusions to be drawn from the work with arrangement A have already been discussed in detail on page 25. Chamber B afforded the possibility of measuring the inset potentials for ionization of argon, neon, krypton, and xenon by argon neutrals. The existence of ionization in all of these cases is believed to be conclusively demonstrated. The check with Varney's value for argon in argon makes this work directly comparable with his in comparing the variation of inset potential with change of particles. His results are given in parentheses in table 1, while those of this work are given without parentheses. It is interesting to note that the inset potential rises as we progress from the diagonal, as would be predicted on the simple theory as expounded by Zwicky. The actual value for the inset in the case of argon in neon cannot be taken too seriously, as the rise of the curve is so slow that the true inset may be somewhat lower than the value of 130 volts estimated from the graphs. The value for argon in xenon is considerably better, though it is true that curve b suggests that there may be some effect below the 55 volts assigned to the inset. The apparent effect may, however, be due to the argon background, since the xenon pressure in this case was rather low. The case for argon in krypton is certainly the most clean cut. Here there can be no question that a very marked effect sets in just above 50 volts.

In discussing measurements of inset potentials one of the principle bones of contention is the relative sensitivities of the balanced space charge method and the electrometer methods. If we compare the results obtained by Varney and those obtained by the author, some light is thrown on this question. With Varney's earlier apparatus (p.13) he was able to measure all

TABLE 1

	<u>Target Atom</u>			
	Gas Bombarded			
	NEON	ARGON	KRYPTON	XENON
Bombarding Atom	NEON	(74)		
	ARGON	130	(48) 48	50 55
	KRYPTON		(40)	
	XENON			(35)

of the noble gases in themselves, with the exception of helium in helium. In this apparatus, the small number of slits allowed a very intense beam of neutrals to be employed. As soon as he increased the number of slits, with a consequent diminution in intensity, he was only able to check his results in argon to 5 volts, and could obtain nothing definite for argon in krypton or xenon. The author's apparatus must have had an inherently greater sensitivity than Varney's, since, in spite of the small intensity of the atom beam, Varney's earlier result for argon in argon was reproduced, and ionization was detected for argon in neon, krypton, and xenon. Had the author been able to use beams of the intensity used in Varney's earlier work, it is probable that the results with neon would have been much more satisfactory.

Always there is the question as to whether or not the insets are truly sharp at these values so far above the electron ionization potentials. The results in the first cases tended to support the hypothesis that the inset was not sharp, but the data for argon in krypton certainly favor the hypothesis of sharp insets. If we were to take curve 13 c, we might be tempted to extrapolate back to 52 volts as the sharp inset. However, when a 52 volt point was taken, curve 13 d, it is seen to make the curve swing in to the background instead of cutting it sharply. It is true that the probability of ionization must rise rapidly between 50 and 55 volts, but the data still do not preclude the possibility of some ionization even at much lower potentials. In order to gain further information on this point it would be enlightening to work with a system in which the beam was homogenised by a good velocity filter, which in turn would remove all of the radiation. The problem of intensity could probably be met by using a vacuum tube electrometer for measuring the ionization. This might also permit the possibility of making measurements of some of the other combinations, particularly some of those using neon as a bombarding agent.

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